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23. ABSTRACT (Continue on reverse if necessary and identify by block number) We have now measured second harmonic generation (1.06 micron source) in DNA films as a function of water content, and counter ion species. The OHG is weak, and does not vary systematically with water content or counter ion species. We have also measured the linear optical properties of DNA at high pressure, finding that electronic properties are very sensitive to pressure, a finding of importance in understanding the electronic basis of imaging of DNA in a scanning tunneling microscope.	
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ANNUAL PROGRESS REPORT**GRANT No. N00014-87-K-0478****RT CODE 441 c 025****PRINCIPAL INVESTIGATOR: S.M. LINDSAY****INSTITUTE: ARIZONA STATE UNIVERSITY****GRANT TITLE: OPTICAL NONLINEARITY IN DNA FILMS****PERIOD OF PERFORMANCE: June 1, 1989 – June 1, 1990****OBJECTIVE:**

To study the linear and nonlinear optical properties of DNA films during a one year, no-cost extension of the original grant (which funded the construction of a nonlinear optics laboratory).

ACCOMPLISHMENTS (last 12 months)

We have now carried out a number of semi-quantitative measurements of optical harmonic generation (OHG) in DNA films with various water and counter-ion contents. The second harmonic signals are generally quite weak, and too variable to yield results which give insight into the mechanism of OHG. Indeed, the main feature of our measurements is the large point to point or sample to sample variation in OHG which does not correlate with obvious optical blemishes or geometrical variations. The films are not phase-matchable, and the sample to sample variations preclude application of the usual powder techniques (e.g. S. Kurtz and T. Perry, J. Appl. Phys., **39** 3798, 1968). However, we can qualitatively characterize our films as having non-linear optical coefficients of the same order of magnitude as crystalline quartz. Some of our data are summarized in the accompanying figure.

We have also investigated the linear optical properties of DNA films at high pressure, with the goal of understanding the changes in electronic properties that might be induced by the contact pressure of a scanning probe, such as that used in the scanning tunneling microscope (STM) or atomic force microscope (AFM). Our initial studies indicate GPa pressures in the STM, enough to shift the molecular orbitals (associated with the bases) by ~ 1 eV and broaden these states by ~ 0.05 eV. Much of this effect can be accounted for by the densification of the DNA. This alters the dielectric constant 'seen' by a molecular orbital in the molecule (this work is described in J. Phys. Chem., **94**, 4655, 1990).

SIGNIFICANCE

Despite the complexity of OHG in DNA films, our data do suggest that they belong to a large class of fibrous biopolymeric materials *which polarize spontaneously*. In all these materials, the origin of the polarization remains a mystery (see, for example, "Optical Second Harmonic Scattering in Rat-tail Tendon" S. Roth and I. Freund, Biopolymers **20**, 1271-1290 (1981)).

Our studies of pressure effects on the linear optical properties suggests a line of investigation now being pursued in our STM/AFM lab. If we can elucidate the electronic basis of tunneling in biopolymers, we will contribute to the interpretation of STM images, and develop a probe of the electronic properties *of individual molecules*.

WORK PLAN

This program is now terminated, and replaced with the STM/AFM work described in an accompanying report.



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INVENTIONS

ASU has filed a patent application for a method of sequencing DNA which relies on contrast in an STM image. An application for a patent on improved electrochemical deposition of molecular adsorbates for scanning probe microscopy is being prepared.

RELEVANT PUBLICATIONS, Last 12 months

"Structure of the DNA Hydration Shells Studied by Raman Spectroscopy" N.J. Tao, S.M. Lindsay and A. Rupprecht, *Biopolymers* **28**, 1019 - 1030 (1989).

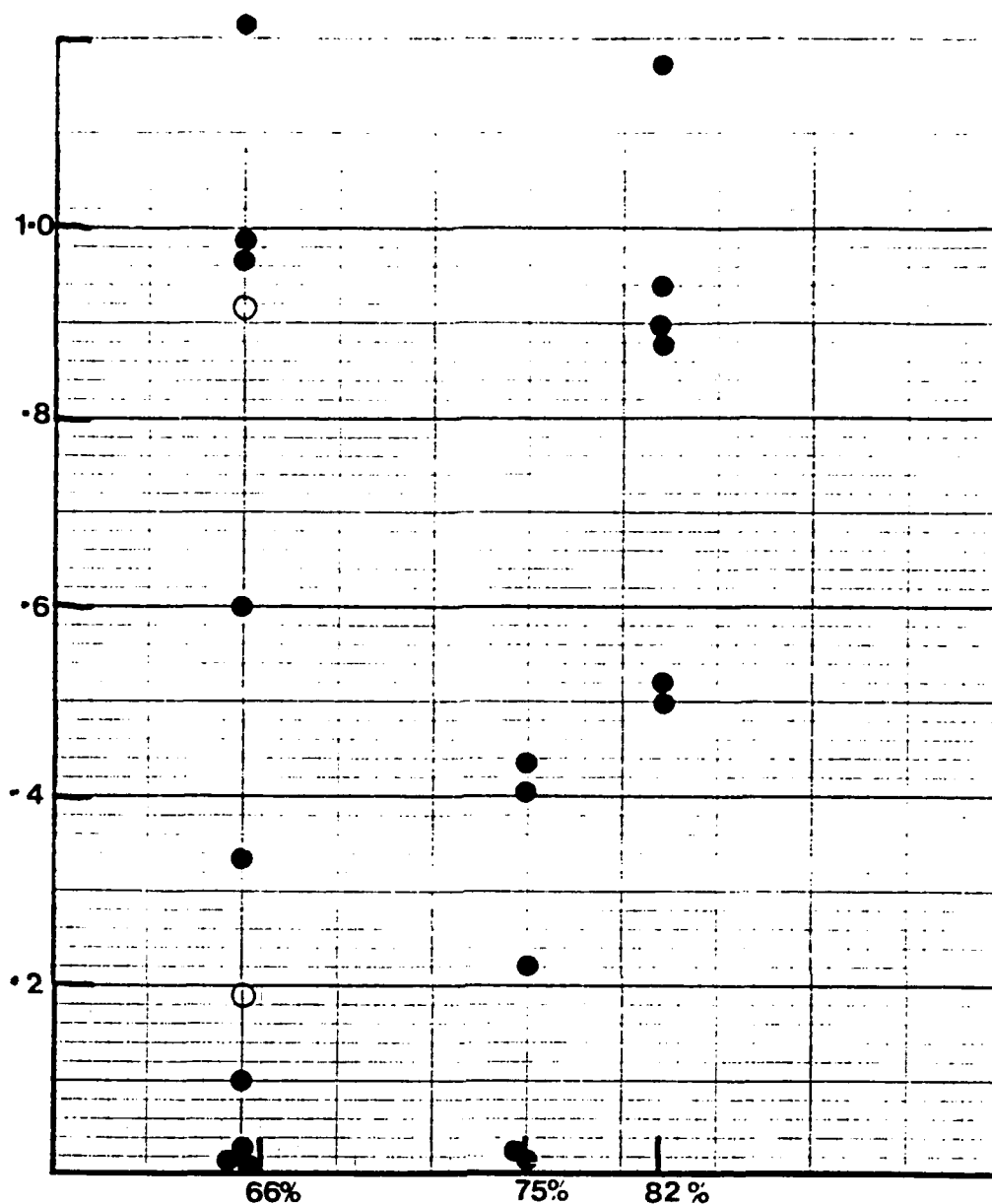
"Reorientational Relaxation of Water Molecules in LiCl Solution Studied by Depolarized Rayleigh Scattering" N.J. Tao and S.M. Lindsay, *J. Phys. Condens. Matter* **1** 8709-8720 (1989).

"Conformational Flexibility in DNA: Ion Mediated Interactions and the Shape of Genes" S.M. Lindsay, *J. Molecular Liquids*, **41** 315-325 (1989).

"Low Frequency Raman Spectra of Z-DNA" T. Weidlich, S.M. Lindsay, W.L. Peticolas and G.A. Thomas, *J. Biomol. Struct. and Dyn.*, in press (1990).

"Pressure and Resonance Effects in Scanning Tunneling Microscopy of Molecular Adsorbates" S.M. Lindsay, O.F. Sankey, Y. Li and C. Herbst, *J. Phys. Chem.*, **94**, 4655 - 4660 (1990).

SECOND HARMONIC GENERATION



RELATIVE HUMIDITY

Figure

Scatter plot of the second harmonic generation versus relative humidity for DNA films. Data points labeled • are for Na-DNA, while those labeled o are for Li-DNA. The scale is arbitrary, but normalized to the OHG from a piece of crystalline quartz maintained in a fixed geometry. Each point is for a different film. However, similar large variations were observed from point to point on a given film.

OBJECTIVES

- Measure Optical Harmonic Generation in DNA films as a function of water content and counter ion.
- Measure linear optical properties to understand electronic states.

ACCOMPLISHMENTS

- Study of range of water contents and two counter ion species. OHG does not depend on these factors in a simple way.
- Electronic states of DNA are very sensitive to pressure. This is important in STM imaging.

SIGNIFICANCE

- DNA films, may, like many other fibrous biopolymers, polarize spontaneously for reasons which remain mysterious.
- Understanding of the electronic properties that permit STM imaging could lead to a probe of the electronic properties of individual molecules.

S.M. Lindsay - Arizona State University

ANNUAL REPORT QUESTIONNAIRE

Principal Investigator: S.M. Lindsay

Institute: Arizona State University

Grant title: Optical Nonlinearity in DNA Films

Period of performance: June 1, 1989 to June 1, 1990

Number of publications last year: 6

Number of patents/inventions: 2

Total number of students/trainees: 2

How many are female? 0

How many are minority students (e.g. Black, Hispanic)? 0

How many are not US citizens? 2

Awards/Honors to PI and/or to members of PI's research group (please describe):

A total of 6 invited talks were given on parts of this work.

Equipment purchased (# and description of items >\$1500):

None

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This address can be reached via Arpanet or Bitnet. We read our mail daily.